

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

LEE et al.

Art Unit: Unassigned

Application No. Unassigned

Examiner: Unassigned

Filed: February 12, 2002

For: TRANSPARENT CONDUCTIVE LAYER
AND IMAGE DISPLAY DEVICE
EMPLOYING THE SAME

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

Prior to the examination of the above-identified patent application, please enter the following amendments and consider the following remarks.

IN THE SPECIFICATION:

Replace the paragraph beginning at page 2, line 4 with:

However, the thin conductive layer formed of the coating composition containing the metal particles has a high grain boundary resistance due to interaction between the particles stabilized by the organic stabilizer, so the surface resistance of the thin conductive layer cannot be lowered. Therefore, there is a need to destroy and remove the organic stabilizer by heating the conductive layer at a high temperature of about 400°C after being coated.

Replace the paragraph beginning at page 4, line 13 with:

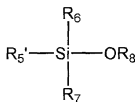
at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

Replace the paragraph beginning at page 6, line 17 with:

at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

Replace the paragraph beginning at page 6, line 22 with:

n is an integer from 0 to 20,



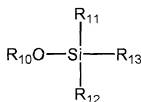
... (2)

where R_5' is a fluorinated $\text{C}_1\text{-C}_{20}$ alkyl group; R_6 and R_7 are, independently, a $\text{C}_1\text{-C}_{20}$ alkoxy group or a fluorinated $\text{C}_1\text{-C}_{20}$ alkyl group; and R_8 is a $\text{C}_1\text{-C}_{20}$ alkyl group,



... (3)

where R_9 is a $\text{C}_1\text{-C}_{20}$ alkyl group, a $\text{C}_1\text{-C}_{20}$ alkyl group with a hydroxy group, a $\text{C}_1\text{-C}_{20}$ hydroxyalkyl group with a hydroxy substituent, or $-(\text{CH}_2)_k\text{COOH}$, where k is an integer from 1 to 10, and



... (4)

where R_{10} is a $\text{C}_1\text{-C}_{20}$ alkyl group; R_{11} and R_{12} are, independently, a $\text{C}_1\text{-C}_{20}$ alkyl group, a $\text{C}_1\text{-C}_{20}$ alkoxy group, or a $\text{C}_1\text{-C}_{20}$ alkyl group with a mercapto group; and R_{13} is a $\text{C}_1\text{-C}_{20}$ alkyl group with a mercapto ($-\text{SH}$) group.

Replace the paragraph beginning at page 7, line 20 with:

Suitable examples of the mercapto compound of formula (3) or (4) above for the spray-coated layer composition are the same as those for the protective layer composition. Preferably, in the spray-coated layer composition, the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of formula (1) above.

Replace the paragraph beginning at page 9, line 6 with:

at least one of R_3 and R_3' is a $\text{C}_1\text{-C}_{20}$ alkoxy group, and the remaining group is a $\text{C}_1\text{-C}_{20}$ alkyl group, a $\text{C}_1\text{-C}_{20}$ alkoxy group, a $\text{C}_2\text{-C}_{20}$ alkylene group, or a $\text{C}_6\text{-C}_{20}$ aryl group;

Amendment to the paragraph beginning at page 10, line 6:

It is preferable that the transparent conductive layer further comprises, on the protective layer, a spray-coated layer containing a hydrolyzed and polycondensated product

of the metal compound of formula (1) above, at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of formula (3) or (4) above and its hydrolyzed and polycondensated product:



where R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group.

Replace the paragraph beginning at page 11, line 15 with:

at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

Replace the paragraph beginning at page 11, line 20 with:

n is an integer from 0 to 20,



where R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group,



where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(\text{CH}_2)_k\text{COOH}$, where k is an integer from 1 to 10, and



where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

Replace the paragraph beginning at page 14, line 4 with:

In formula (1) above, M is selected from the group consisting of Si, Ti, Sn, and Zr; R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group; R_2 is a C_1 - C_{20} alkyl group; at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and n is an integer from 0 to 20,



Replace the paragraph beginning at page 15, line 16 with:

As described above, in formula (1) above of the metal compound, it is preferable that at least one of the groups R_4 and R_5 is a C_1 - C_{20} alkoxy group and at least one of the groups R_3 and R_3' is a C_1 - C_{20} alkoxy group. Such a metal compound with these groups can form a 3-dimensional network structure of metal oxide, such as silica, through hydrolysis and polycondensation.

Replace the paragraph beginning at page 17, line 29 with:

In formula (2) above, R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group.

Replace the paragraph beginning at page 18, line 1 with:

For the groups R_5' , R_6 , R_7 , and R_8 in formula (2) above, suitable examples of the fluorinated C_1 - C_{20} alkyl group include a heptadecafluorodecyl group, a pentadecafluorohexyl group and the like, and suitable examples of the C_1 - C_{20} alkoxy group include a methoxy group, an ethoxy group, a propoxy group, an isopropoxy group, a butoxy group and the like.

Replace the paragraph beginning at page 22, line 11 with:

The spray-coated layer composition may further comprise a hydrolytic catalyst. Nitric acid, hydrochloric acid, phosphoric acid, sulfuric acid and the like can be used as the hydrolytic catalyst. The hydrolytic catalyst may be used in an amount of 0.1-0.9 mole, preferably 0.3-0.7 mole, with respect to 1 mole of the metal compound of formula (1) above.

If a hydrolytic catalyst less than 0.1 mole is used, the manufacturing process may be extended. If a hydrolytic catalyst more than 0.9 mole is used, it may be difficult to control the manufacturing process.

Replace the paragraph beginning at page 23, line 1 with:

In formula (1) above, M is selected from the group consisting of Si, Ti, Sn, and Zr; R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group; R_2 is a C_1 - C_{20} alkyl group; at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and n is an integer from 0 to 20.

Replace the paragraph beginning at page 23, line 12 with:

In manufacturing the first transparent conductive layer, a metal oxide of an average particle size of 5-5000 nm is dispersed in a polar solvent to prepare a conductive layer composition. A top surface of the glass panel is coated with the conductive layer composition and dried at 30-100°C to form a conductive layer. The drying process may be not performed if necessary.

Replace the paragraph beginning at page 23, line 24 with:

A network structure between silica and mercapto compound is formed during calcination, as shown in FIG. 2. This calcination is performed at, preferably a temperature of 100-400°C. If the calcination temperature is above 400°C, the types of available substrates may be limited. If the calcination temperature is less than 100°C, the film hardness may be not strong enough.

Replace the paragraph beginning at page 24, line 29 with:

A transparent conductive layer according to the present invention may additionally include a spray-coated layer 14 on the protective layer 13, as shown in FIG. 1B. To form the spray-coated layer 14, the protective layer 13 is coated with a spray-coated layer composition, which contains a metal compound of formula (1) above, fluoroalkylsilane of formula (2) above, a mercapto compound of formula (3) or (4), and a polar solvent. Next, the resulting structure is dried and calcinated. Drying temperature is in the range of 30-100°C. The drying process may be omitted if necessary. Calcination temperature is in the range of 100-400°C. If the calcination temperature is less than 100°C, the film hardness may be reduced. If the calcination temperature is above 100°C, the image display device may be likely to be broken.

Replace the paragraph beginning at page 25, line 30 with:

In manufacturing the second transparent conductive layer according to the present invention, as in the manufacture of the first transparent conductive layer, a metal oxide of an average particle size of 5-5000 nm is dispersed in a polar solvent to prepare a conductive layer composition. A top surface of a glass panel is coated with the conductive layer composition and dried at 30-100°C to form a conductive layer. The drying process may be not performed if necessary.

Replace the paragraph beginning at page 26, line 3 with:

Separately, a metal compound of formula (1) above is dispersed in a polar solvent to prepare a protective layer composition. The conductive layer is coated with this protective layer composition and dried at 30-100°C to form a protective layer. The drying process may be omitted if necessary, as in formation of the conductive layer.

Replace the paragraph beginning at page 29, line 11 with:

Separately, to a solvent mixture of 50 g of ethanol, 30 g of methanol, 10 g of isopropyl alcohol, and 7.25 g of n-butanol were added 2.67 g of tetraethyl silicate and 0.08 g of mercaptopropyltrimethoxysilane. The mixture was stirred and aged at 60°C for 24 hours to prepare a protective layer composition.

Replace the paragraph beginning at page 29, line 15 with:

The conductive layer composition was spin coated on a substrate, dried, and coated with the protective layer composition. The resulting structure was thermally treated at 200°C for 30 minutes to form a transparent conductive layer.

Replace the paragraph beginning at page 29, line 20 with:

A transparent conductive layer was formed in the same manner as in Example 11, except that the protective layer composition was slightly changed. To a solvent mixture of 50 g of ethanol, 30 g of methanol, 10 g of isopropyl alcohol, and 7.25 g of n-butanol were added 2.67 g of tetraethylorthosilicate and 0.16 g of 3-mercaptopropyltrimethoxysilane. The mixture was stirred and aged at 60°C for 24 hours to prepare a protective layer composition.

Replace the paragraph beginning at page 30, line 11 with:

Film hardness at the surface was determined using a pencil hardness method. Standard pencils with a tip hardness in H were used. The cross-section of each pencil tip was polished to be planar and was moved once on the surface of each conductive layer at a 45° tilt and a speed of 0.5 cm/min with the application of a load of 1 kg to the pencil in a vertical

direction. It was observed whether the surface of the conductive layer was scratched or not. The same test was performed while changing the pencils having different hardnesses. The largest hardness of the pencils at which no scratch was observed for a conductive layer was read as the hardness of the conductive layer.

Replace the paragraph beginning at page 30, line 22 with:

Variation in resistance was observed after the transparent conductive layers were left at 45°C and 95% RH for 24 hours.

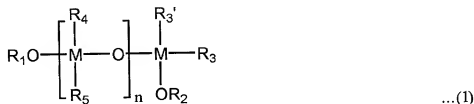
Replace the paragraph beginning at page 30, line 26 with:

After the transparent conductive layers were left at 45°C and 95% RH for 24 hours, the surfaces of the transparent conductive layers were visually observed to determine whether the surfaces were stained or not.

IN THE CLAIMS:

Replace the indicated claims with:

1. (Amended) A protective layer composition comprising a metal compound of formula (1) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group;

R_2 is a C_1 - C_{20} alkyl group;

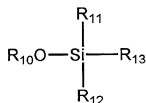
at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and

n is an integer from 0 to 20,



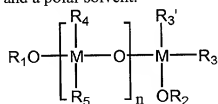
where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



...(4)

where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

8. (Amended) A spray-coated layer composition comprising a metal compound of formula (1) below, fluoroalkylsilane of formula (2) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



...(1)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

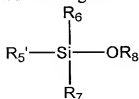
R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group;

R_2 is a C_1 - C_{20} alkyl group;

at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and

n is an integer from 0 to 20,



...(2)

where R_{5'} is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group,

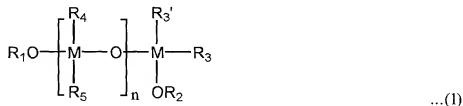
R₉ SH ...(3)

where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_kCOOH, where *k* is an integer from 1 to 10, and



where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

16. (Amended) A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer formed on the conductive layer, the protective layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below and at least one of a mercapto compound of formula (3) or (4) below and its hydrolyzed and polycondensated product:



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;
at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and
 n is an integer from 0 to 20,



where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



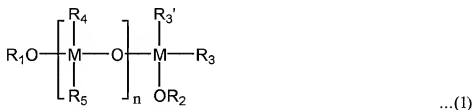
where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

21. (Amended) The transparent conductive layer of claim 16, further comprising, on the protective layer, a spray-coated layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1), at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of said formula (3) or (4) and its hydrolyzed and polycondensated product:



where R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group.

24. (Amended) A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer and spray-coated layer sequentially formed to protect the conductive layer, the spray-coated layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below, at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of formula (3) or (4) below and its hydrolyzed and polycondensated product, and the protective layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1):



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

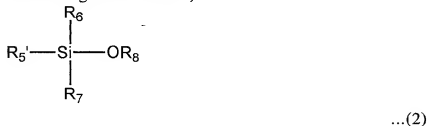
R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

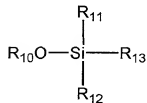
n is an integer from 0 to 20,



where R₃' is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group,



where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_kCOOH, where k is an integer from 1 to 10, and



...(4)

where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

REMARKS

The specification and claims has been amended to remove obvious typographical errors.
No new matter has been added.

If, in the opinion of the Examiner, a telephone conference would expedite the
prosecution of the subject application, the Examiner is invited to call the undersigned attorney.

Respectfully submitted,

LEYDIG, VOIT & MAYER, LTD.



Xavier Pillai, Ph.D.
Registration No. 39,799

Suite 300
700 Thirteenth Street, N.W.
Washington, D.C. 20005
Telephone: (202) 737-6770
Facsimile: (202) 737-6776
Date: 2/12/02
XP:jj

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

LEE et al.

Application No. Unassigned

Art Unit: Unassigned

Filed: February 12, 2002

Examiner: Unassigned

For: TRANSPARENT CONDUCTIVE
LAYER AND IMAGE DISPLAY
DEVICE EMPLOYING THE SAME

AMENDMENTS TO SPECIFICATION AND CLAIMS
MADE VIA PRELIMINARY AMENDMENT

Amendment to the paragraph beginning at page 2, line 4:

However, the thin conductive layer formed of the coating composition containing the metal particles has a high grain boundary resistance due to interaction between the particles stabilized by the organic stabilizer, so the surface resistance of the thin conductive layer cannot be lowered. Therefore, there is a need to destroy and remove the organic stabilizer by heating the conductive layer at a high temperature of about ~~400EC~~ 400°C after being coated.

Amendment to the paragraph beginning at page 4, line 13:

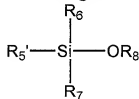
at least one of R₃ and ~~R₃~~ R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

Amendment to the paragraph beginning at page 6, line 17:

at least one of R₃ and ~~R₃~~ R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

Amendment to the paragraph beginning at page 6, line 22:

n is an integer from 0 to 20,



...(2)

where R_4-N-R_5 is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group,



where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

Amendment to the paragraph beginning at page 7, line 20:

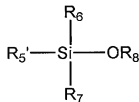
Suitable examples of the ~~mercapto-mercapto~~ compound of formula (3) or (4) above for the spray-coated layer composition are the same as those for the protective layer composition. Preferably, in the spray-coated layer composition, the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of formula (1) above.

Amendment to the paragraph beginning at page 9, line 6:

at least one of R_3 and R_4-N-R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

Amendment to the paragraph beginning at page 10, line 6:

It is preferable that the transparent conductive layer further comprises, on the protective layer, a spray-coated layer containing a hydrolyzed and polycondensated product of the metal compound of formula (1) above, at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of formula (3) or (4) above and its hydrolyzed and polycondensated product:



...(2)

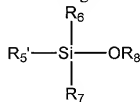
where ~~R₅-N-R₅'~~ is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group.

Amendment to the paragraph beginning at page 11, line 15:

at least one of R₃ and ~~R₃-N-R₃'~~ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

Amendment to the paragraph beginning at page 11, line 20:

n is an integer from 0 to 20,

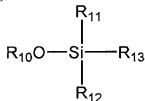


...(2)

where ~~R₅-N-R₅'~~ is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group,

R₉ SH ...(3)

where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_{*k*}COOH, where *k* is an integer from 1 to 10, and



...(4)

where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

Amendment to the paragraph beginning at page 14, line 4:

In formula (1) above, M is selected from the group consisting of Si, Ti, Sn, and Zr; R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group; R₂ is a C₁-C₂₀ alkyl group; at least one of R₃ and R₃-N-R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and n is an integer from 0 to 20,



Amendment to the paragraph beginning at page 15, line 16:

As described above, in formula (1) above of the metal compound, it is preferable that at least one of the groups R₄ and R₅ is a C₁-C₂₀ alkoxy group and at least one of the groups R₃ and R₃-N-R₃' is a C₁-C₂₀ alkoxy group. Such a metal compound with these groups can form a 3-dimensional network structure of metal oxide, such as silica, through hydrolysis and polycondensation.

Amendment to the paragraph beginning at page 17, line 29:

In formula (2) above, R₆-N-R₆' is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group.

Amendment to the paragraph beginning at page 18, line 1:

For the groups R₆-N-R₆', R₆, R₇, and R₈ in formula (2) above, suitable examples of the fluorinated C₁-C₂₀ alkyl group include a heptafluorodecyl group, a pentafluorohexyl group and the like, and suitable examples of the C₁-C₂₀ alkoxy group include a methoxy group, an ethoxy group, a propoxy group, an isopropoxy group, a butoxy group and the like.

Amendment to the paragraph beginning at page 22, line 11:

The spray-coated layer composition may further comprise a hydrolytic catalyst. Nitric acid, hydrochloric acid, phosphoric acid, sulfuric acid and the like can be used as the hydrolytic catalyst. The hydrolytic catalyst may be used in an amount of 0.1-0.9 mole, preferably 0.3-0.7 mole, with respect to 1 mole of the metal compound of formula (1) above. If a hydrolytic catalyst less than 0.1 mole is used, the manufacturing process may be extended. If a hydrolytic catalyst more than 0.9 mole is used, it may be difficult to control the manufacturing process.

Amendment to the paragraph beginning at page 23, line 1:

In formula (1) above, M is selected from the group consisting of Si, Ti, Sn, and Zr; R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group; R₂ is a C₁-C₂₀ alkyl group; at least one of R₃ and R₄ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and n is an integer from 0 to 20.

Amendment to the paragraph beginning at page 23, line 12:

In manufacturing the first transparent conductive layer, a metal oxide of an average particle size of 5-5000 nm is dispersed in a polar solvent to prepare a conductive layer composition. A top surface of the glass panel is coated with the conductive layer composition and dried at ~~30-100°C~~ 30-100°C to form a conductive layer. The drying process may be not performed if necessary.

Amendments to the paragraph beginning at page 23, line 24:

A network structure between silica and mercapto compound is formed during calcination, as shown in FIG. 2. This calcination is performed at, preferably a temperature of ~~100-400°C~~ 100-400°C. If the calcination temperature is above ~~400°C~~ 400°C, the types of available substrates may be limited. If the calcination temperature is less than ~~100°C~~ 100°C, the film hardness may be not strong enough.

Amendments to the paragraph beginning at page 24, line 29:

A transparent conductive layer according to the present invention may additionally include a spray-coated layer 14 on the protective layer 13, as shown in FIG. 1B. To form the spray-coated layer 14, the protective layer 13 is coated with a spray-coated layer composition, which contains a metal compound of formula (1) above, fluoroalkylsilane of formula (2) above, a mercapto compound of formula (3) or (4), and a polar solvent. Next, the resulting structure is dried and calcinated. Drying temperature is in the range of ~~30-100°C~~ 30-100°C. The drying process may be omitted if necessary. Calcination temperature is in the range of ~~100-400°C~~ 100-400°C. If the calcination temperature is less than ~~100°C~~ 100°C, the film hardness may be reduced. If the calcination temperature is above ~~100°C~~ 100°C, the image display device may be likely to be broken.

Amendment to the paragraph beginning at page 25, line 30:

In manufacturing the second transparent conductive layer according to the present invention, as in the manufacture of the first transparent conductive layer, a metal oxide of an average particle size of 5-5000 nm is dispersed in a polar solvent to prepare a conductive layer composition. A top surface of a glass panel is coated with the conductive layer composition and dried at ~~30-100°C~~ 30-100°C to form a conductive layer. The drying process may be not performed if necessary.

Amendment to the paragraph beginning at page 26, line 3:

Separately, a metal compound of formula (1) above is dispersed in a polar solvent to prepare a protective layer composition. The conductive layer is coated with this protective layer composition and dried at ~~30-100°C~~ 30-100°C to form a protective layer. The drying process may be omitted if necessary, as in formation of the conductive layer.

Amendment to the paragraph beginning at page 29, line 11:

Separately, to a solvent mixture of 50 g of ethanol, 30 g of methanol, 10 g of isopropyl alcohol, and 7.25 g of n-butanol were added 2.67 g of tetraethyl silicate and 0.08 g of mercaptopropyltrimethoxysilane. The mixture was stirred and aged at ~~60°C~~ 60°C for 24 hours to prepare a protective layer composition.

Amendment to the paragraph beginning at page 29, line 15:

The conductive layer composition was spin coated on a substrate, dried, and coated with the protective layer composition. The resulting structure was thermally treated at ~~200°C~~ 200°C for 30 minutes to form a transparent conductive layer.

Amendment to the paragraph beginning at page 29, line 20:

A transparent conductive layer was formed in the same manner as in Example 11, except that the protective layer composition was slightly changed. To a solvent mixture of 50 g of ethanol, 30 g of methanol, 10 g of isopropyl alcohol, and 7.25 g of n-butanol were added 2.67 g of tetraethylorthosilicate and 0.16 g of 3-mercaptopropyltrimethoxysilane. The mixture was stirred and aged at ~~60°C~~ 60°C for 24 hours to prepare a protective layer composition.

Amendment to the paragraph beginning at page 30, line 11:

Film hardness at the surface was determined using a pencil hardness method. Standard pencils with a tip hardness in H were used. The cross-section of each pencil tip was polished to be planar and was moved once on the surface of each conductive layer at a ~~45°~~ 45° tilt and a speed of 0.5 cm/min with the application of a load of 1 kg to the pencil in a

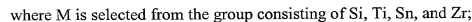
Amendment to the paragraph beginning at page 30, line 22:

Amendment to the paragraph beginning at page 30, line 26:

After the transparent conductive layers were left at 45°C and 95% RH for 24 hours, the surfaces of the transparent conductive layers were visually observed to determine whether the surfaces were stained or not.

Amendments to existing claims:

1. (Amended) A protective layer composition comprising a metal compound of formula (1) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

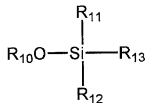
at least one of R₃ and R_{3-N-R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;}

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkenylene group, or a C₆-C₂₀ aryl group; and

n is an integer from 0 to 20,

$$R_0 SH \quad \dots (3)$$

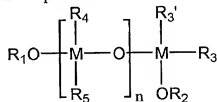
where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



...(4)

where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

8. (Amended) A spray-coated layer composition comprising a metal compound of formula (1) below, fluoroalkylsilane of formula (2) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



...(1)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

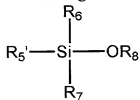
R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group;

R_2 is a C_1 - C_{20} alkyl group;

at least one of R_3 and R_3' is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and

n is an integer from 0 to 20,



...(2)

where R_5-N-R_6 is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group,

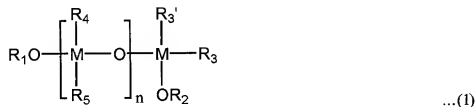


where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

16. (Amended) A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer formed on the conductive layer, the protective layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below and at least one of a mercapto compound of formula (3) or (4) below and its hydrolyzed and polycondensated product:



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R_1 is a C_1 - C_{20} alkyl group or $-M(R_{14}R_{15}R_{16})$ where R_{14} , R_{15} , and R_{16} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_6 - C_{20} aryl group;

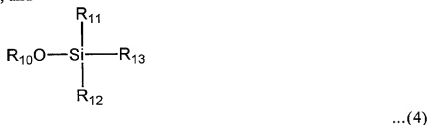
R_2 is a C_1 - C_{20} alkyl group;

at least one of R_3 and R_3-N-R_8 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group;

at least one of R_4 and R_5 is a C_1 - C_{20} alkoxy group, and the remaining group is a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkylene group, or a C_6 - C_{20} aryl group; and
 n is an integer from 0 to 20,



where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(CH_2)_kCOOH$, where k is an integer from 1 to 10, and



where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

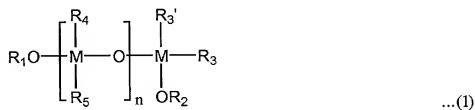
21. (Amended) The transparent conductive layer of claim 16, further comprising, on the protective layer, a spray-coated layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1), at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of said formula (3) or (4) and its hydrolyzed and polycondensated product:



where R_5 and R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group.

24. (Amended) A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer and spray-coated layer sequentially formed to protect the

conductive layer, the spray-coated layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below, at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of formula (3) or (4) below and its hydrolyzed and polycondensated product, and the protective layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1):



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

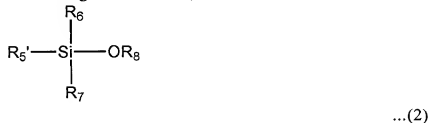
R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

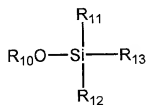
n is an integer from 0 to 20,



where R₃-R₃' is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group,



where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_kCOOH, where k is an integer from 1 to 10, and



...(4)

where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

LEE et al.

Application No. Unassigned

Art Unit: Unassigned

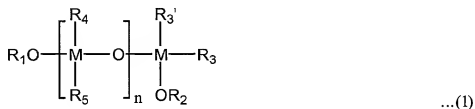
Filed: February 12, 2002

Examiner: Unassigned

For: TRANSPARENT CONDUCTIVE
LAYER AND IMAGE DISPLAY
DEVICE EMPLOYING THE SAME

PENDING CLAIMS AFTER ENTRY OF PRELIMINARY AMENDMENT

1. A protective layer composition comprising a metal compound of formula (1) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

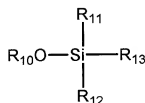
at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

n is an integer from 0 to 20,



where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_kCOOH, where k is an integer from 1 to 10, and



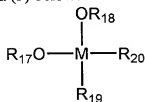
...(4)

where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

2. The protective layer composition of claim 1, wherein the mercapto compound of said formula (3) or (4) is at least one selected from the group consisting of 3-mercaptopropyltrimethoxysilane, 3-mercaptopropylmethyldimethoxysilane, 3-mercaptopropyl-1,2-propanediol, 1-mercaptopropyl-2-propanol, 3-mercaptopropionic acid, di-(3-mercaptopropyl)dimethoxysilane, and tris-(3-mercaptopropyl)methoxysilane, and the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

3. The protective layer composition of claim 1, wherein the metal compound of said formula (1) is at least one selected from the group consisting of tetraethylorthosilicate, tetramethylorthosilicate, methyltrimethoxyorthosilicate, vinyltriethoxysilane, 3-glycidypropyltrimethoxysilane, and phenyltriethoxysilane.

4. The protective layer composition of claim 1, further comprising a metal compound of formula (5) below:



...(5)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁₇ and R₁₈ are, independently, a C₁-C₂₀ alkyl group or a C₆-C₂₀ aryl group; and

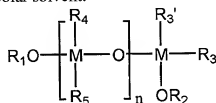
R₁₉ and R₂₀ are, independently, a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group.

5. The protective layer composition of claim 4, wherein the metal compound of said formula (5) is at least one selected from the group consisting of dimethyldimethoxyorthosilicate, diethyldimethoxyorthosilicate, dimethyldiethoxyorthosilicate, and diethyldiethoxyorthosilicate.

6. The protective layer composition of claim 1, wherein the polar solvent is at least one selected from the group consisting of ethanol, methanol, butanol, isopropanol, methylethylketone, methylcellosolve, and ethylcellosolve, and the polar solvent is contained in an amount of 1000-4000 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

7. The protective layer composition of claim 1, further comprising a hydrolytic catalyst in an amount of 0.1-0.9 mole with respect to 1 mole of the metal compound of said formula (1), and the hydrolytic catalyst is at least one selected from the group consisting of nitric acid, hydrochloric acid, phosphoric acid, and sulfuric acid.

8. A spray-coated layer composition comprising a metal compound of formula (1) below, fluoroalkylsilane of formula (2) below, a mercapto compound of formula (3) or (4) below, and a polar solvent:



...(1)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

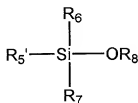
R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R₃ and R_{3'} is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

n is an integer from 0 to 20,



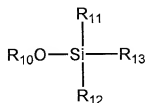
...(2)

where R_5' is a fluorinated C_1 - C_{20} alkyl group; R_6 and R_7 are, independently, a C_1 - C_{20} alkoxy group or a fluorinated C_1 - C_{20} alkyl group; and R_8 is a C_1 - C_{20} alkyl group,



...(3)

where R_9 is a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkyl group with a hydroxy group, a C_1 - C_{20} hydroxyalkyl group with a hydroxy substituent, or $-(\text{CH}_2)_k\text{COOH}$, where k is an integer from 1 to 10, and



...(4)

where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto ($-\text{SH}$) group.

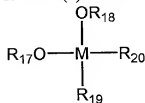
9. The spray-coated layer composition of claim 8, wherein the fluoroalkylsilane of said formula (2) is at least one selected from the group consisting of heptadecafluorodecyltriethoxysilane, pentadecafluorohexyltrimethoxysilane, heptadecafluorodecyltrimethoxysilane, heptadecafluorodecyltriisopropoxysilane, heptadecafluorodecyltributoxysilane, di-(heptadecafluorodecyl)diethoxysilane, and tris-(heptadecafluorodecyl)ethoxysilane, and the fluoroalkylsilane of said formula (2) is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of formula (1).

10. The spray-coated layer composition of claim 8, wherein the mercapto compound of said formula (3) or (4) is at least one selected from the group consisting of 3-mercaptopropyltrimethoxysilane, 3-mercaptopropylmethyldimethoxysilane, 3-mercapto-1,2-

propanediol, 1-mecapto-2-propanol, 3-mercaptopropionic acid, di-(3-mercaptopropyl)dimethoxysilane, and tris-(3-mercaptopropyl)methoxysilane, and the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

11. The spray-coated layer composition of claim 8, wherein the metal compound of said formula (1) is at least one selected from the group consisting of tetraethylorthosilicate, tetramethylorthosilicate, methyltrimethoxyorthosilicate, vinyltriethoxysilane, 3-glycidoxypropyltrimethoxysilane, and phenyltriethoxysilane.

12. The spray coated layer composition of claim 8, further comprising a metal compound of formula (5) below:



...(5)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁₇ and R₁₈ are, independently, a C₁-C₂₀ alkyl group or a C₆-C₂₀ aryl group; and

R₁₉ and R₂₀ are, independently, a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group.

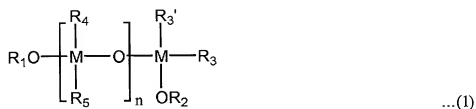
13. The spray-coated layer composition of claim 12, wherein the metal compound of said formula (5) is at least one selected from the group consisting of dimethyldimethoxyorthosilicate, diethyldimethoxyorthosilicate, dimethylethoxyorthosilicate, and diethyldiethoxyorthosilicate.

14. The spray-coated layer composition of claim 8, wherein the polar solvent is at least one selected from the group consisting of ethanol, methanol, butanol, isopropanol, methylethylketone, methylcellosolve, and ethylcellosolve, and the polar solvent is contained in an amount of 1000-4000 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

15. The spray-coated layer composition of claim 9, further comprising a hydrolytic catalyst in an amount of 0.1-0.9 mole with respect to 1 mole of the metal compound of said formula (1),

and the hydrolytic catalyst is at least one selected from the group consisting of nitric acid, hydrochloric acid, phosphoric acid, and sulfuric acid.

16. A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer formed on the conductive layer, the protective layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below and at least one of a mercapto compound of formula (3) or (4) below and its hydrolyzed and polycondensated product:



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R₃ and R_{3'} is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

n is an integer from 0 to 20,



where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_{*k*}COOH, where *k* is an integer from 1 to 10, and

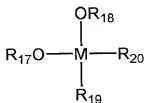


where R₁₀ is a C₁-C₂₀ alkyl group; R₁₁ and R₁₂ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₁-C₂₀ alkyl group with a mercapto group; and R₁₃ is a C₁-C₂₀ alkyl group with a mercapto (-SH) group.

17. The transparent conductive layer of claim 16, wherein the mercapto compound of said formula (3) or (4) is at least one selected from the group consisting of 3-mercaptopropyltrimethoxysilane, 3-mercaptopropylmethyldimethoxysilane, 3-mercapto-1,2-propanediol, 1-mercapto-2-propanol, 3-mercaptopropionic acid, di-(3-mercaptopropyl)dimethoxysilane, and tris-(3-mercaptopropyl)methoxysilane, and the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

18. The transparent conductive layer of claim 16, wherein the metal compound of said formula (1) is at least one selected from the group consisting of tetraethylorthosilicate, tetramethylorthosilicate, methyltrimethoxyorthosilicate, vinyltriethoxysilane, 3-glycidoxypropyltrimethoxysilane, and phenyltriethoxysilane.

19. The transparent conductive layer of claim 16, wherein the protective layer further contains a hydrolyzed and polycondensated product of a metal compound of formula (5) below:



...(5)

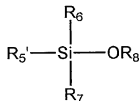
where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁₇ and R₁₈ are, independently, a C₁-C₂₀ alkyl group or a C₆-C₂₀ aryl group; and

R₁₉ and R₂₀ are, independently, a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group.

20. The transparent conductive layer of claim 19, wherein the metal compound of said formula (5) is at least one selected from the group consisting of dimethyldimethoxyorthosilicate, diethyldimethoxyorthosilicate, dimethyldiethoxyorthosilicate, and diethyldiethoxyorthosilicate.

21. The transparent conductive layer of claim 16, further comprising, on the protective layer, a spray-coated layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1), at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of said formula (3) or (4) and its hydrolyzed and polycondensated product:

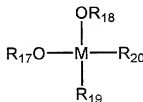


...(2)

where R_5' is a fluorinated $\text{C}_1\text{-C}_{20}$ alkyl group; R_6 and R_7 are, independently, a $\text{C}_1\text{-C}_{20}$ alkoxy group or a fluorinated $\text{C}_1\text{-C}_{20}$ alkyl group; and R_8 is a $\text{C}_1\text{-C}_{20}$ alkyl group.

22. The transparent conductive layer of claim 21, wherein the spray-coated layer is formed as a non-continuous layer.

23. The transparent conductive layer of claim 21, wherein the spray-coated layer further contains a hydrolyzed and polycondensated product of a metal compound of formula (5) below:



...(5)

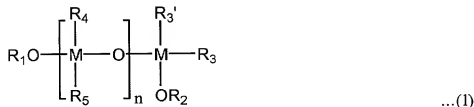
where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R_{17} and R_{18} are, independently, a $\text{C}_1\text{-C}_{20}$ alkyl group or a $\text{C}_6\text{-C}_{20}$ aryl group; and

R_{19} and R_{20} are, independently, a $\text{C}_1\text{-C}_{20}$ alkyl group, a $\text{C}_2\text{-C}_{20}$ alkylene group, or a $\text{C}_6\text{-C}_{20}$ aryl group.

24. A transparent conductive layer comprising a conductive layer containing a metal oxide and a protective layer and spray-coated layer sequentially formed to protect the conductive layer, the spray-coated layer containing a hydrolyzed and polycondensated product of a metal compound of formula (1) below, at least one of fluoroalkylsilane of formula (2) below and its hydrolyzed and polycondensated product, and at least one of a mercapto compound of

formula (3) or (4) below and its hydrolyzed and polycondensated product, and the protective layer containing a hydrolyzed and polycondensated product of the metal compound of said formula (1):



where M is selected from the group consisting of Si, Ti, Sn, and Zr;

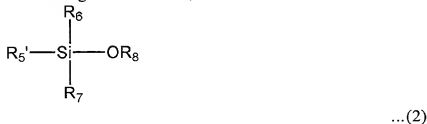
R₁ is a C₁-C₂₀ alkyl group or -M(R₁₄R₁₅R₁₆) where R₁₄, R₁₅, and R₁₆ are, independently, a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, or a C₆-C₂₀ aryl group;

R₂ is a C₁-C₂₀ alkyl group;

at least one of R₃ and R₃' is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkoxy group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group;

at least one of R₄ and R₅ is a C₁-C₂₀ alkoxy group, and the remaining group is a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group; and

n is an integer from 0 to 20,



where R₅' is a fluorinated C₁-C₂₀ alkyl group; R₆ and R₇ are, independently, a C₁-C₂₀ alkoxy group or a fluorinated C₁-C₂₀ alkyl group; and R₈ is a C₁-C₂₀ alkyl group,



where R₉ is a C₁-C₂₀ alkyl group, a C₁-C₂₀ alkyl group with a hydroxy group, a C₁-C₂₀ hydroxyalkyl group with a hydroxy substituent, or -(CH₂)_kCOOH, where k is an integer from 1 to 10, and



where R_{10} is a C_1 - C_{20} alkyl group; R_{11} and R_{12} are, independently, a C_1 - C_{20} alkyl group, a C_1 - C_{20} alkoxy group, or a C_1 - C_{20} alkyl group with a mercapto group; and R_{13} is a C_1 - C_{20} alkyl group with a mercapto (-SH) group.

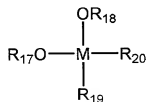
25. The transparent conductive layer of claim 24, wherein the spray-coated layer is formed as a non-continuous layer.

26. The transparent conductive layer of claim 24, wherein the fluoroalkylsilane of said formula (2) is at least one selected from the group consisting of heptadecafluorodecyltriethoxysilane, pentadecafluorohexyltrimethoxysilane, heptadecafluorodecyltrimethoxysilane, heptadecafluorodecyltriisopropoxysilane, heptadecafluorodecyltributoxysilane, di-(heptadecafluorodecyl)diethoxysilane, and tris-(heptadecafluorodecyl)ethoxysilane, and the fluoroalkylsilane of said formula (2) is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of formula (1).

27. The transparent conductive layer of claim 24, wherein the mercapto compound of said formula (3) or (4) is at least one selected from the group consisting of 3-mercaptopropyltrimethoxysilane, 3-mercaptopropylmethyldimethoxysilane, 3-mercapto-1,2-propanediol, 1-mercapto-2-propanol, 3-mercaptopropionic acid, di-(3-mercaptopropyl)dimethoxysilane, and tris-(3-mercaptopropyl)methoxysilane, and the mercapto compound is contained in an amount of 1-15 parts by weight based on 100 parts by weight of the metal compound of said formula (1).

28. The transparent conductive layer of claim 24, wherein the metal compound of said formula (1) is at least one selected from the group consisting of tetraethylorthosilicate, tetramethylorthosilicate, methyltrimethoxyorthosilicate, 3-glycidoxypropyltrimethoxysilane, vinyltriethoxysilane, and phenyltriethoxysilane.

29. The transparent conductive layer of claim 24, wherein the spray-coated layer further contains a hydrolyzed and polycondensated product of a metal compound of formula (5) below:



...(5)

where M is selected from the group consisting of Si, Ti, Sn, and Zr;

R₁₇ and R₁₈ are, independently, a C₁-C₂₀ alkyl group or a C₆-C₂₀ aryl group; and

R₁₉ and R₂₀ are, independently, a C₁-C₂₀ alkyl group, a C₂-C₂₀ alkylene group, or a C₆-C₂₀ aryl group.

30. The transparent conductive layer of claim 29, wherein the metal compound of said formula (5) is at least one selected from the group consisting of dimethyldimethoxyorthosilicate, diethyldimethoxyorthosilicate, dimethylethoxyorthosilicate, and diethyldiethoxyorthosilicate.

31. An image display device employing the transparent conductive layer of claim 16.

32. The image display device of claim 31, wherein the transparent conductive layer is formed on a panel of a cathode ray tube.

33. An image display device employing the transparent conductive layer of claim 24.

34. The image display device of claim 33, wherein the transparent conductive layer is formed on a panel of a cathode ray tube.

Amendment - Preliminary (Rev. 7/5/2001)